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THE ZONE MELTING OF TUNGSTEN BY ELECTRON BOMBARDMENT

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As part of the NASA's general program of research on materials for use at very high temperatures, we have, at the Lewis Research Center, a substantial effort on the metallurgy of tungsten and tungsten alloys. Since it is generally believed that the brittle behavior of recrystallized tungsten is caused by small concentrations of impurity elements, a portion of this research is devoted to producing high purity tungsten and to determining if the elimination of impurities from tungsten significantly increases its ductility.

In order to reduce the impurity content in commercially available tungsten we have employed floating-zone melting using electron bombardment for heating. The important advantages of this method are a high temperature capability, a high vacuum condition, and a low power requirement.

The purposes of this paper is to describe the apparatus used for the zone melting of tungsten rods, to indicate some of the problems involved, and to discuss some of the results.

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APPARATUS

A photograph of the electron bombardment zone melting unit is shown in figure 1. The unit consists of a vacuum chamber containing the zone melting mechanism, assorted vacuum pumping equipment, and the electrical power and control equipment.

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The vacuum chamber is approximately 24 inches high by 10 inches in diameter and is made of standard forged steel pipe. A water jacket surrounds the chamber. Several flanged openings sealed with O-ring gaskets are provided for a sight port and for vacuum connections.

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The chamber is evacuated by an Evapor-ion (titanium-getter) pump and a six-inch diffusion pump package. A liquid nitrogen trap is used in conjunction with the oil diffusion pump to minimize backstreaming of oil into the melting chamber.

As can be seen in figure 2, the zone-melting mechanism is attached to the base-plate of the vacuum chamber. This mechanism is made up of a number of small components: the specimen, the specimen support, the electron "gun," a lead screw and the gun carriage, and two electrical leads. The specimen remains stationary while the gun traverses up and down. The lead screw providing motion to the gun carriage is driven by a small d.c. motor through a gear reducer and a rotary vacuum seal. The copper electrical leads pass through and are insulated from the base plate by Mycalex insulators. In order to prevent a short circuit at this location due to coating of the insulators with evaporated tungsten, the insulators are shielded by Pyrex tubes.

The electron gun consists of a 0.020 inch diameter tungsten filament surrounded by several molybdenum focusing plates. The filament, made in the form of a loop about 3/4 inch in diameter, is spaced approximately 1/4 inch from the top and bottom focusing plates. The specimen passes through a 3/8 inch opening in the center of the plates. An outer shield of molybdenum is included to aid in focusing the electrons. A small opening in this shield permits the operator to see the molten zone. Molybdenum screws hold the filament in place and also act as terminal posts for the flexible copper leads.

A functional block diagram illustrating the electrical circuit used for electron bombardment melting is shown in figure 3. High voltage d.c.

power is supplied by a transformer and two rectifier tubes. This supply of power is limited in voltage by a Variac and in current by a series of resistors. The high voltage is impressed on the filament, thus driving the electrons emitted by the heated filament to the grounded specimen. Current used in the electron bombardment is sensed by a resistor. The voltage drop across the resistor is compared to a control voltage determined by a variable resistor and battery circuit. The voltage difference or error is fed into the filament heat control, in this case, a Brown amplifier that drives the Variac feeding the filament transformer. The feedback, thereby, controls the electron emission and the bombardment current by varying the filament temperature.

OPERATION

The procedure normally followed in zone melting a 1/8-inch diameter tungsten rod is to initially outgas the rod at a power level just below that required for producing surface melting. About 400 watts are presently used. With the gun traveling at about 10 mm/min at this power, the zone temperature is to about 5000° F. The melting pass that follows is usually carried out at a slower gun traverse rate, about 2.5 mm/min, in a downward direction and at a power level of about 600 watts. The power level is adjusted according to the appearance of the floating zone. If a bulge is seen in the bombarded zone, the core of the tungsten rod is considered to be completely molten. Subsequent metallographic examination of the zone-melted rods showed that the structure was uniform from the surface to the center, thus confirming that the rods had been completely melted. If too much power is applied, the surface tension forces become insufficient to support the molten zone and the rod "burns through."

The melting operation is usually carried out at a pressure of approximately 5×10^{-6} mm Hg. To avoid carbon contamination from backstreaming diffusion pump oil the Evapor-ion pump is used alone. The pressure can be further lowered to about 2×10^{-6} mm Hg during melting by using the additional pumping capacity of the 6-inch diffusion pump with its liquid nitrogen trap.

A number of difficulties have been experienced during melting. The most common problem is an overcurrent condition resulting from sudden gas bursts that can't be compensated for by the controller or the current limiter. An overcurrent relay breaks the circuit when this condition occurs. The melting can usually be restarted without much noticeable difference in the uniformity of the specimen diameter.

In order to provide the maximum amount of zone-refined material for evaluation, we would like to produce zone-melted rods of uniform diameter. However, the bulge formed during melting carries away a small amount of tungsten from the starting region, leaving a necked section, and deposits this amount at the end of the pass. A rod having only these imperfections is shown in figure 4.

About 50 percent of the tungsten rods have a fairly uniform diameter after the first melting pass. The remainder have some defect. Often the rod will have a series of necked portions instead of the single initial necking that should take place. The cause of this rippled appearance is not known, but is believed to be related to such factors as the sharp temperature gradient existing in the heated zone and the vibration from pumps and other equipment. Further surface irregularity on the rod is attributed to control instability resulting from the outgassing of the rod and the deposition of vapors on the filament loop.

RESULTS

Up to this time, most of our effort has been devoted to developing the zone melting apparatus. We have, however, successfully zone melted both 1/8- and 3/16-inch diameter tungsten rods. An attempt was made to zone melt 1/4-inch tungsten rods but the power available was sufficient only for surface melting.

In order to achieve maximum purification of tungsten by zone refining, we would like to be able to make multiple melting passes. Although rods of relatively uniform diameter have been obtained after repeated melting, a defect on the rods in one pass may often be emphasized in the succeeding pass. Because of the desire to evaluate the zone melted material produced in this equipment, we have thus far concentrated on rods given only a single melting pass.

X-ray diffraction photographs of the edge of rotating specimens of 1/8- and 3/16-inch tungsten rods indicate that a single crystal is usually formed during the first zone-melting pass. Macroetching of zone-melted rods also indicate that they are single crystals. Certain crystallographic planes are selectively etched and extend the full length of the zone-melted section.

We have not yet adequately determined the extent of purification achieved by the zone-melting of tungsten in our apparatus. However, the few available chemical analyses are of interest in that they indicate that a single melting pass significantly lowers the impurity content of commercially pure tungsten. For example, the oxygen content, initially at 140 ppm, was brough down to 4 ppm; iron was reduced from 40 ppm to less than 10 ppm; and molybdenum from 80 ppm to less than 30 ppm.

In order to determine if zone-refining improves the ductility of tungsten, we have conducted room temperature bend tests. The results of some of the bend tests are illustrated in figure 5. The "as received" (swaged and centerless ground) rod exhibited no bend ductility. Another piece of this rod, which had been recrystallized during an annealing pass in an attempt to degas it, was also brittle. In contrast, the one-pass zone-melted rod was quite ductile and bent through an angle of 110° without fracture. The test fixture which has a fulcrum with a 3/32-inch radius limited the angle of bend to 110°. It is not known at this time whether the increased ductility is the result of purification achieved by zone-melting or is solely due to the fact that the zone-melted rod is a single crystal.

A few high temperature tensile tests of the zone-melted tungsten rods have been conducted. Figure 6 shows the chisel-shaped fracture of a specimen that was evaluated at 2500° F. Slip lines can be seen along the length of the specimen where deformation occurred.

Swaging attempts at room temperature resulted in shattering of the zone-melted material but swaging at 1600° to 1700° F was readily accomplished. The zone-melted rod also rolled easily when heated to about 900° F.

CONCLUDING REMARKS

The electron bombardment zone-refining apparatus described in this paper has proved useful for preparing high purity tungsten single crystals. Modifications are being made to the equipment in an attempt to improve the surface uniformity of the zone-melted rods. We are currently attempting to determine the effect of multiple melting passes on the purification of tungsten, utilizing both chemical analyses and low temperature electrical resistivity measurements to establish impurity concentrations. In this way, we hope to establish a better correlation between tungsten purity and its mechanical properties.

Figure 1

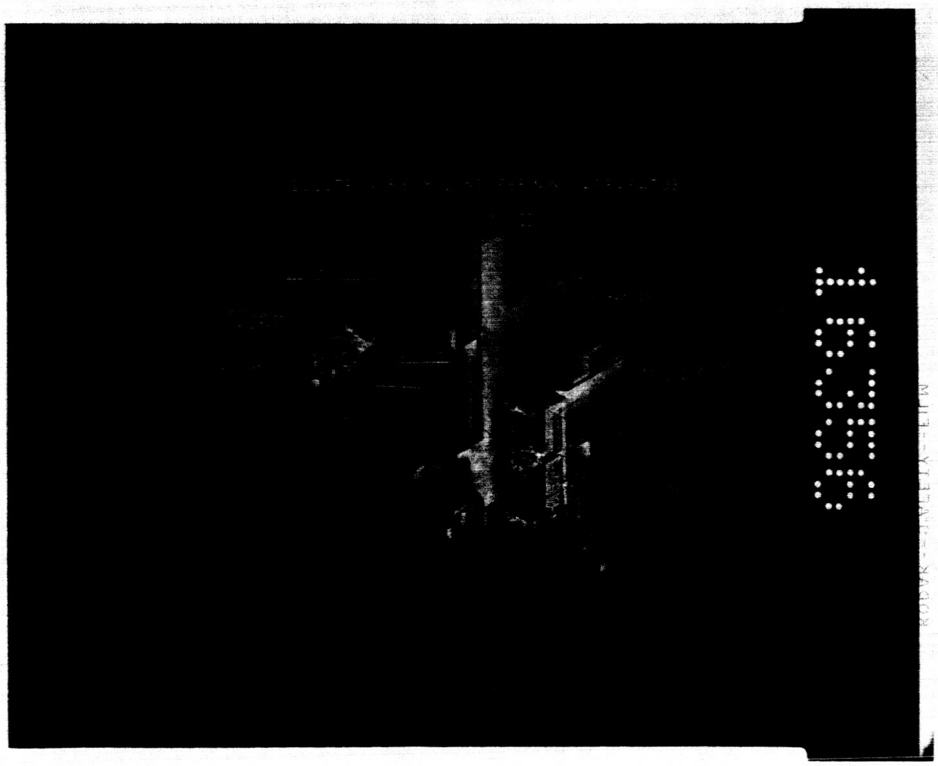

Figure 2

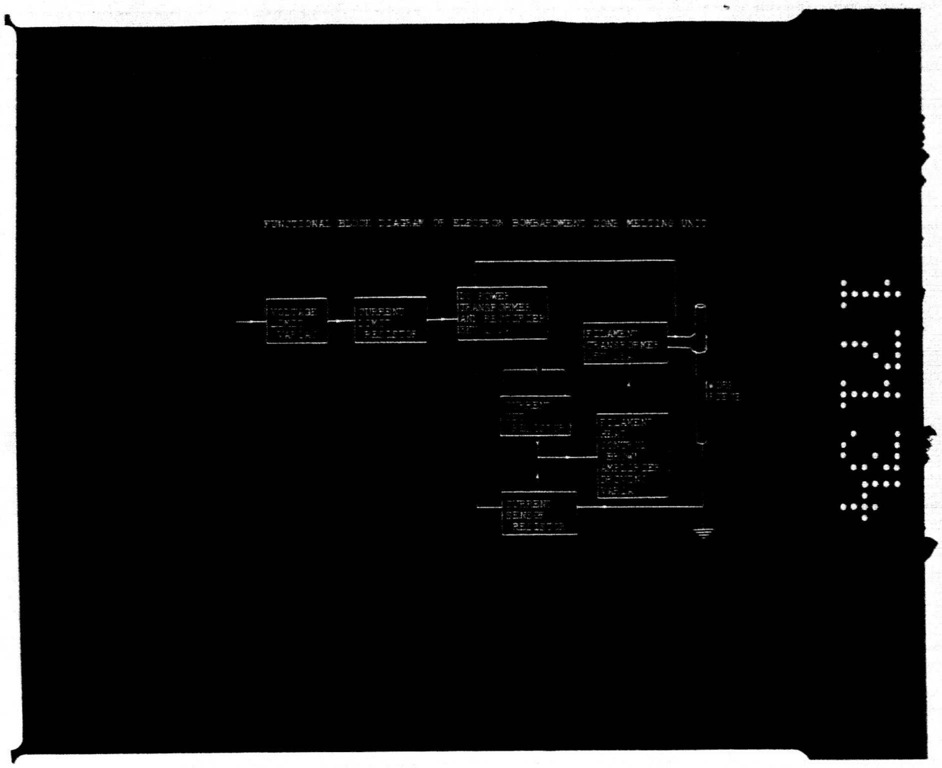

Figure 3

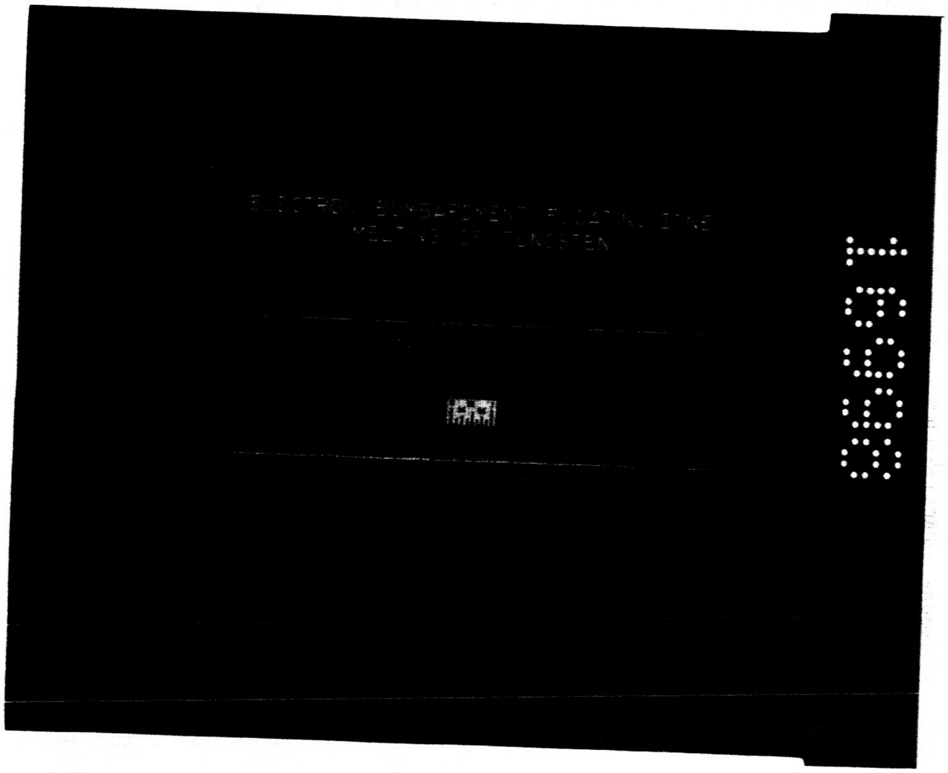

Figure 4

Figure 5

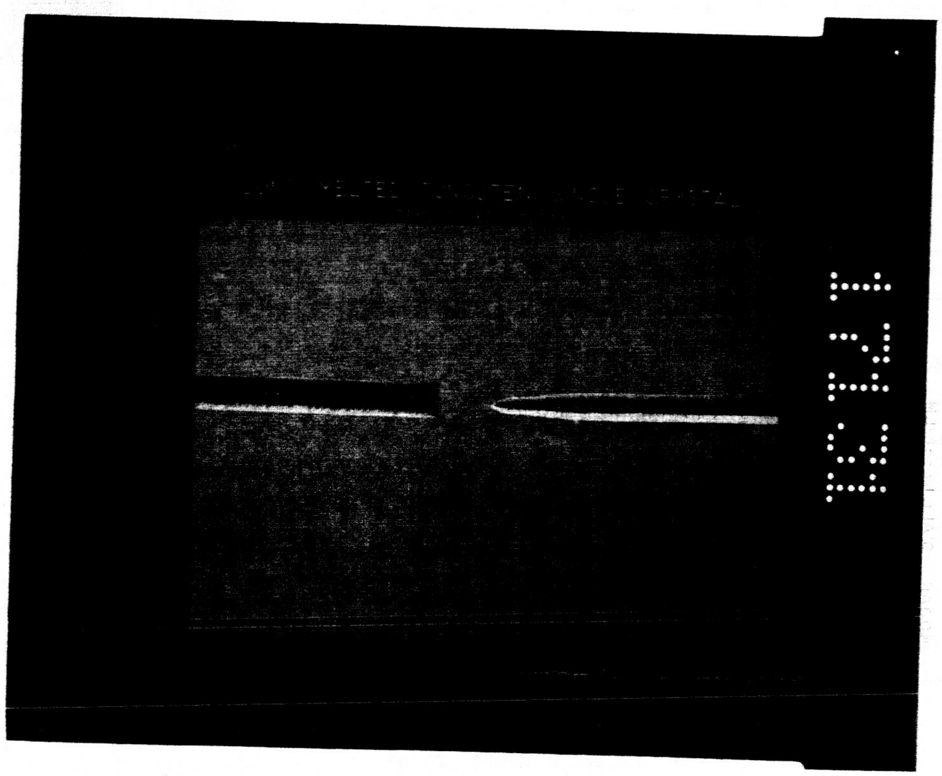

Figure 6